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Controlling the Induced Anisotropy in Soft Magnetic Films for High-Frequency Applications

A. R. Chezan, C. B. Craus, N. G. Chechenin, T. Vystavel, J. Th. M. De Hosson, L. Niesen, and D. O. Boerma

***Abstract*—Nanocrystalline soft magnetic Fe–Zr–N films were successfully deposited by dc magnetron reactive sputtering. The nitrogen content was controlled by varying the Ar/N_2 ratio and/or the substrate temperature. The films have saturation magnetization and induced uniaxial anisotropy values in the range 17–21 kG and 0–30 Oe, respectively. The origin of the induced anisotropy is discussed. The films can be used as magnetic cores for inductors at gigahertz frequencies.**

***Index Terms*—Fe–N films, high-frequency properties, nanocrystalline materials, soft magnetic materials, uniaxial anisotropy.**

I. Introduction

SOFT magnetic materials with controllable uniaxial anisotropy are required for future ultrahigh-frequency applications. Currently, the attention is focused on Fe–M–N (M = Ta [1], Cr [2], etc.) systems due to their high saturation magnetization (M_S), excellent magnetic softness, and good corrosion resistance. The materials must present a well-controlled domain pattern with the magnetization, in the absence of an external magnetic field, oriented parallel to the direction of the anisotropy axis. The hysteresis loop will then be a rectangle in the easy direction (domain wall movement) and a straight line in the hard direction (spin rotation). When an external ac field is applied perpendicular to the easy axis the magnetization oscillates with the field. The permeability in this case is given by the ratio between the M_S and the anisotropy field (H_K). For thin films, where the eddy current losses can be neglected, this value can be maintained up to the ferromagnetic resonance (FMR) frequency, which is proportional to $(M_S H_K)^{1/2}$ [2]. In order to push this limit into the gigahertz range, films with high M_S and H_K in excess of 20 Oe are desired.

The magnetic domain pattern can be controlled by reduction of the grain size below the ferromagnetic exchange length (35 nm in Fe) [3], [4] and by introducing a uniaxial anisotropy. The films can be obtained in an As-deposited nanocrystalline state by sputtering Fe-rich alloys in a reactive atmosphere (Ar+N_2) [5]. The anisotropy is created by a bias magnetic field applied during deposition or during a subsequent magnetic annealing treatment. In the past decade, much effort was spent on optimizing the microstructure and composition of these materials in order to get optimal magnetic properties. However, the information about the origin and magnitude of the uniaxial anisotropy is often contradictory. In this paper, we describe a method for fine tuning the structural and magnetic properties of sputtered films, and we discuss the origin of the induced anisotropy in Fe–Zr–N films.

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II. Experiment

Fe–Zr–N films with a thickness between 50 and 200 nm were grown in a nanocrystalline structural state on Si wafers by dc magnetron reactive sputtering. Fe sheets partially covered with Zr wires were used as targets, giving a ratio Fe/Zr in the films of 99/1 or 97.5/2.5. The nitrogenation was controlled by varying the substrate temperature during deposition and/or the Ar/N_2 (7 or 18 vol% N_2) gas mixture. The combinations of target composition and sputtering mixture used are indicated in the legend of the figures. An 800-Oe magnetic field was applied in the plane of the samples during production. The thickness and composition of the films were measured by Rutherford backscattering spectrometry (RBS) and elastic recoil detection (ERD). The phases, texture, lattice spacing, and nitrogen content were determined by standard X-ray diffraction (XRD) and by two-dimensional (2-D) XRD texture measurements. Transmission electron microscopy (TEM) was used for structure analysis. Hysteresis loops were recorded using a vibrating sample magnetometer (VSM). The anisotropy field was determined using the method described in [6].

III. Results and Discussions

Nitrogen is the key component of the Fe–M–N thin films for obtaining excellent soft magnetic properties. In order to preserve relatively high values of the M_S, the films should be composed mainly by an Fe-rich bcc phase with N dissolved interstitially. This phase is known as α' Fe nitride. The lattice structure of the α' phase has been the subject of many investigations. The N atoms are located at the interstitial octahedral sites of the bcc matrix, with four neighboring Fe atoms in the (100) plane and two Fe atoms in the [100] direction, the octahedron axis, perpendicular to the (100) plane. This octahedron is too small to contain an N atom, and some lattice distortion is required for accommodation. In pure Fe, it has been found that the lattice is uniformly dilated for N contents below 2.7 at% [7]. For higher N contents, a transition to a tetragonal distorted phase with the c axis along the [100] direction is observed. The c/a ratio increases linearly with the N content, while the value of a diminishes very slightly with increasing N content. It was found that the transition point of 2.7 at% can be shifted to lower values by applying uniaxial stress on the sample [8]. The spontaneous

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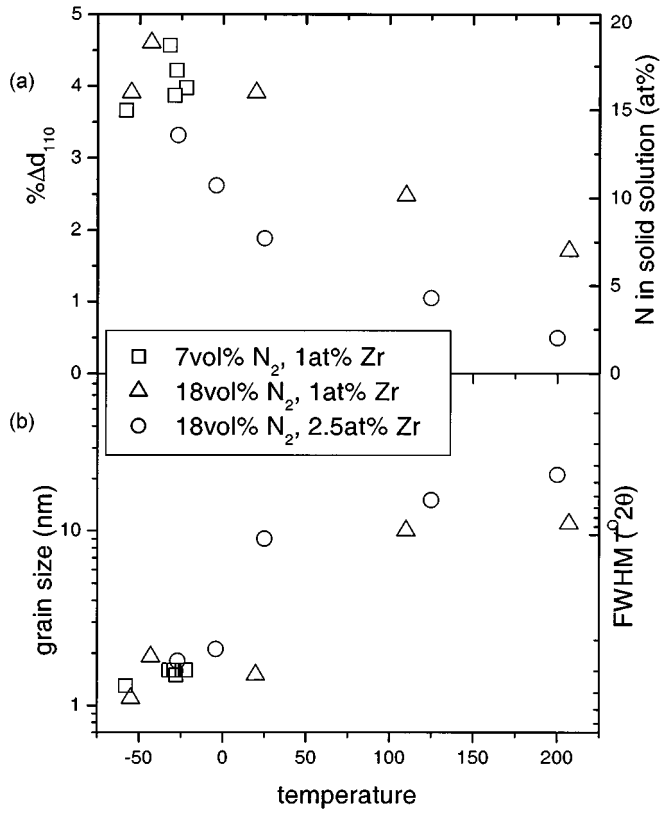

Fig. 1. Influence of the deposition temperature on (a) the relative variation of the corrected lattices parameter and the N concentration, and (b) the broadening of the X-ray diffraction line and the corresponding average grain size.

cubic-to-tetragonal ordering can also be pushed to higher N concentrations [9].

All the Fe–Zr–N films investigated in this study present a {110} fibrous texture with the fiber axis perpendicular to the plane of the films. Standard XRD scans show a shift to lower 2θ values and a broadening of the bcc (110) reflection as the deposition temperature decreases (Fig. 1). We took great care to measure with 2-D XRD whether in our samples a tetragonal lattice distortion is present. We found that the [110] interplanar distance is equal for sets of planes parallel to the surface and for sets of planes making an angle of 60° with the surface. The conclusion is that for samples with grain size larger than 10 nm, a uniform cubic lattice dilation occurs for any N concentration. For samples with smaller grains, the XRD diffraction peaks are strongly broadened by the Scherrer effect. For this reason, a tetragonal distortion for samples with very small grains cannot be detected.

The shift of the diffraction line is given in Fig. 1(a) as a relative variation of the [110] interplanar distance (Δd_{110}) corrected for the influence of Zr, which was assumed to be in solid solution in the Fe matrix. The corresponding lattice dilation can be correlated with the quantity of N in interstitial solid solution in the bcc matrix. For values of the N content between 3 and 9 at%, the corrected lattice dilation is proportional to the N content [8]. Deviations from this behavior are found for samples deposited at 200 °C, suggesting that at this temperature only a fraction of the N contributes to the lattice dilation. The rest of the N is probably bound to Zr, forming small ZrN clusters. At

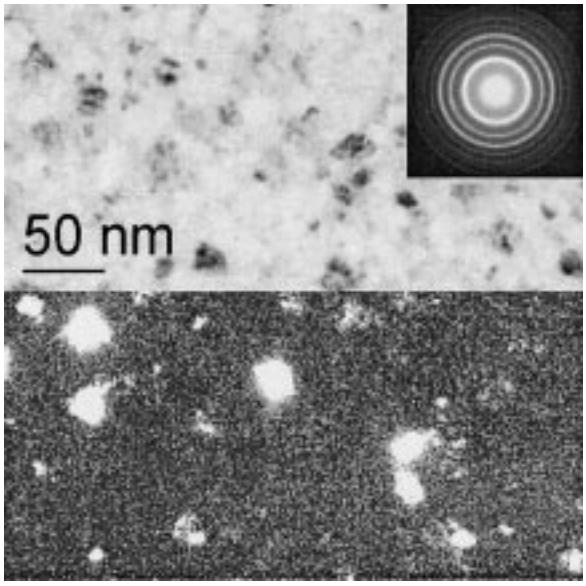

Fig. 2. (a) Planar-view BF and (inset) SAD pattern and (b) DF images of a film deposited at 200 °C from a target with an Fe/Zr atomic ratio of 97.5/2.5 and in a sputtering gas containing 18 vol% N_2.

lower temperatures, most N atoms are in solid solution in the Fe matrix, as suggested by the corresponding lattice dilation. Reducing the deposition temperature below room temperature (RT) lead to an incorporation of nitrogen in the films above the 11 at% limit of α' while no traces of other Fe–N compounds (γ', ε) were found [Fig. 1(a)].

The average grain size was estimated from the width of the same bcc (110) diffraction line using the Scherrer relation [10]. In this estimation, we ignored other sources of line broadening such as inhomogeneous stress fields. However, by cross checking with TEM results, we learned that the use of the Scherrer relation leads to a good estimate of the grain size.

In Fig. 2, planar-view bright field (BF) and dark field (DF) images of the same area of a 50-nm-thick film deposited at 200 °C are shown. The BF image [Fig. 2(a)] has a complex contrast, typical for nanocrystalline materials with grain sizes smaller than the film thickness and due to the presence of defects inside the grains. In the DF image [Fig. 2(b)], crystallites with sizes in the 5–20-nm range become visible. The irregular boundaries of the crystallites suggest the existence of transition regions between the grains composed of distorted material. The selected area diffraction (SAD) pattern presented in the inset of Fig. 2(a) shows that there is no in-plane preferred orientation of the crystallites. Cross-section DF images using the main (110) reflection (not shown) reveal a clear columnar structure of the films. This columnar structure plays an important role in promoting a perpendicular anisotropy component, for film thickness above 200 nm, having as a result the disappearance of the soft magnetic behavior. Below RT, the films have an average grain size smaller than 2 nm, and further decreasing the deposition temperature to −50 °C has little influence on this value [Fig. 1(b)].

The values of the M_S of the Fe–Zr–N films range from 21 to 17 kG as the deposition temperature decreases. In epitaxial or polycrystalline (with micrometer-sized grains) Fe–N films, the

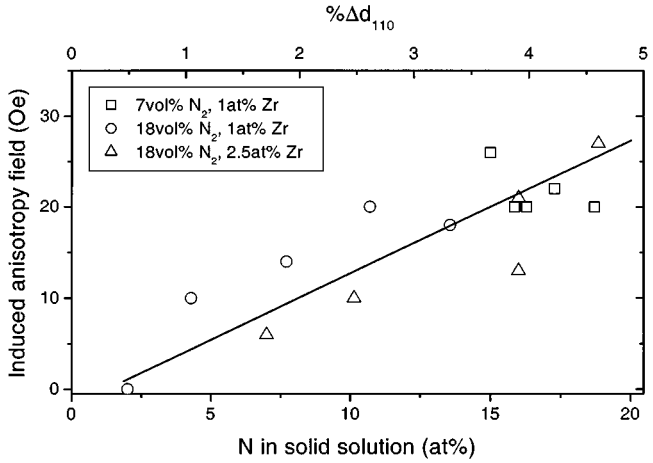

Fig. 3. Variation of the induced uniaxial anisotropy as a function of the interstitial N.

increase of the N content between 0 and 11 at% leads to an increase of M_S from 21 kG corresponding to pure Fe to approximately 26 kG corresponding to the "famous" α'' phase [11]. In the case of the Fe–N or Fe–M–N nanocrystalline systems, in all reports known by the authors, in the same interval of N concentrations M_S decreases with the N content. Our findings are in agreement with the latter observations. Possible explanations for this effect are the distorted intergranular material with a lower M_S [1] and the presence of Zr and N as nonmagnetic impurities in the Fe matrix.

The microscopic source of uniaxial magnetic anisotropy in Fe–M–N materials is the tetragonal symmetry of the N containing octahedron. The overall induced uniaxial anisotropy of the material suggests that the N atoms occupy sites such that the [100] axes of the octahedra are aligned [12].

The induced uniaxial anisotropy increases linearly with the interstitial nitrogen content in the sputtered Fe–Zr–N films, as suggested by Fig. 3. The Zr–N pairs, formed due to the high affinity between the Zr and N atoms, play a minor role in the induced anisotropy since materials with 1 at% and, respectively, 2.5 at% Zr present practically the same value for the anisotropy at the same values of the N content in the films. As stated earlier, our extensive investigations show that in the nanocrystalline Fe–Zr–N alloys, a tetragonal lattice distortion is not present at concentrations of N as high as 8 at%. The lattice is uniformly dilated. A possible explanation for this experimental fact can be that due to the small size of the grains the driving force for the cubic to tetragonal transition is too small to produce the phase transformation. The quantity of N atoms within a grain is insufficient to produce a tetragonal distortion even if they are all located in octahedra pointing in the same crystallographic orientation. At the same time, the films containing 8 at% N present a maximum induced magnetic anisotropy of 10 Oe, which can be changed both in magnitude and direction by magnetic annealing at temperatures < 250 °C. At these low temperatures, only the nitrogen atoms are mobile. This provides another argument that the magnetic anisotropy is not caused by, or accompanied by, a large tetragonal distortion, which should cause large strains in the films. It is not to be expected that such a redirection of a significant tetragonal distortion could take place at this temperature. The maximum value of the induced anisotropy is proportional to the quantity of N atoms dissolved interstitially in the iron matrix, and the proportionality coefficient in our case (≈ 1 Oe/at% N) is similar to that reported in [12]. This last observation suggests that the large majority of the N atoms contribute to the induced anisotropy. Therefore, our conclusion is that in the Fe–Zr–N nanocrystalline films, the N atoms can be aligned without producing a tetragonal distortion. Irrespective of the particular arrangement of the N atoms, the grains are dilated isotropically. The proportionality between the N content and the induced uniaxial magnetic anisotropy offers a modality of fine tuning the soft magnetic properties of nanocrystalline Fe–Zr–N films.

According to the high-frequency measurements (an example is given in [13]), the nanocrystalline Fe–Zr–N films with an induced anisotropy in excess of 25 Oe present a flat response up to 1.8 GHz.

IV. Conclusion

It has been shown that the deposition conditions, temperature, sputtering mixture, and growth rate, offer the possibility of controlling the soft magnetic properties of Fe–Zr–N films. Due to the "grain refinement" action of N, nanocrystalline films can be obtained in the as-deposited state. This effect is amplified by lowering the temperature of the substrate during deposition. The induced anisotropy originates in the alignment of the majority of the interstitial N. The alignment of N atoms is realized without producing a tetragonal distortion of the bcc Fe lattice. Zr–N pairs do not contribute significantly to the induced anisotropy. The films are promising candidates for ultrahigh-frequency applications.

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